The demand must be filed directly with	h the competent International Preliminary Examining Authority or, if two or more Authorities are competent The full name or two-letter code of that Authority may be indicated by the applicant on the line below:
with the one chosen by the applicant.	The full name or two-letter code of that Authority may be that the of the applicable of the and octors.

IPEA/	 <del></del>	 	

## **PCT**

**CHAPTER II** 

## **DEMAND**

under Article 31 of the Patent Cooperation Treaty:

The undersigned requests that the international application specified below be the subject of international preliminary examination according to the Patent Cooperation Treaty.

Identification of IPEA		Date of receipt of DEMAND		
Box No. I DENTIFICATION OF THE INTERNATIONAL		APPLICATION	Applicant's or agent's file reference DCJ131PCT	
nternational application No. PCT/JP2004/005014	International filing date 07/04/2		(Earliest) Priority date (day/month/year) 07/04/2003	
Title of invention Curable Organopolysiloxane R Fransmission Components, an	esin Composition for design of the control of the c	or Optical Trans	mission Components, Optical	
Box No. II APPLICANT(S)		•	*	
Name and address: (Family name followed by The address must include p		full official designation.	Telephone No. 0465-76-3108	
DOW CORNING ASIA LTD. 1-3, Marunouchi 1-chome, C	Chiyoda-ku, Tokyo	)	Facsimile No. 0465-76-3308	
1000005 Japan			Teleprinter No.	
			Applicant's registration No. with the Office 000109185	
State (that is, country) of nationality:  JAPAN		State (that is, coun	ntry) of residence:	
KUSHIBIKI, Nobuo 2-6-22 Honfujisawa Fujisaw 2510875 Japan			ne address must include postal code and name of country	
State (that is, country) of nationality: JAPAN		State (that is, cour JAPAN	ntry) of residence:	
		full official designation. T	The address must include postal code and name of country	
State (that is, country) of nationality:		State (that is, coun	ntry) of residence:	

Sheet No. .2.

International application No. PCT/JP2004/005014

Continuation of Box No. II APPLICANT(S)					
If none of the following sub-boxes is used, this sheet should not be included in the demand.					
Name and address: (Family name followed by given name: for a legal entity, full TAKEUCHI, Kikuko 57-2 Ikoma Minamiashigara-shi Kanagawa 2500124 Japan	official designation. The address must include postal code and name of country.)				
State (that is, country) of nationality: JAPAN	State (that is, country) of residence: JAPAN				
Name and address: (Family name followed by given name; for a legal entity, fu	Il official designation. The address must include postal code and name of country.)				
State (that is, country) of nationality:	State (that is, country) of residence:				
Name and address: (Family name followed by given name: for a legal entity, ful	l official designation. The address must include postal code and name of country.)				
State (that is, country) of nationality:	State (that is, country) of residence:				
Name and address: (Family name followed by given name; for a legal entity, fu					
State (that is, country) of nationality:  State (that is, country) of residence:					
Further applicants are indicated on another continuation sheet.					

Sheet No. . 3.

International application No. PCT/JP2004/005014

BOX NO. III AGENT OR COMMON REPRESENTATIVE; OR ADDRESS FOR CORRESPONDENCE					
The following person is agent common representative					
and X has been appointed earlier and represents the applicant(s) also for international preliminary examination.					
is hereby appointed and any earlier appointment of (an) agent(s)/common represer	tative is hereby revoked.				
is hereby appointed, specifically for the procedure before the International Prelimithe agent(s)/common representative appointed earlier.	nary Examining Authority, in addition to				
Name and address: (Family name followed by given name; for a legal entity, full official designation.  The address must include postal code and name of country.)	Telephone No.				
	81-043-216-6030				
Patent Attorney	Facsimile No.				
KUBOTA, Yoshitaka c/o Asa International Patent Firm Chiba Office	81-043-216-6030 Teleprinter No.				
43-27, Satsukigaoka 1-chome,	relepinioi ivo.				
Hanamigawaku,	Agent's registration No. with the Office				
Chiba-shi, Chiba 262-0014 JAPAN	100091579				
Address for correspondence: Mark this check-box where no agent or common a space above is used instead to indicate a special address to which correspondence	epresentative is/has been appointed and the should be sent.				
Box No. IV BASIS FOR INTERNATIONAL PRELIMINARY EXAMINATION					
Statement concerning amendments:*					
1. The applicant wishes the international preliminary examination to start on the basis of	:				
the international application as originally filed	1				
the description as originally filed					
as amended under Article 34					
the claims as originally filed	ag statement)				
as amended under Article 19 (together with any accompanying	ng statement)				
as amended under Article 34					
the drawings as originally filed					
as amended under Article 34					
2. The applicant wishes any amendment to the claims under Article 19 to be considered as reversed.					
The applicant wishes the start of the international preliminary examination to be postponed until the expiration of the					
applicable time limit under Rule 69.1(d).  4. The applicant expressly wishes the international preliminary examination to start earlier than at the expiration of the					
applicable time limit under Rule 54bis. 1(a).					
* Where no check-box is marked, international preliminary examination will start on the basis of the international application as originally filed or, where a copy of amendments to the claims under Article 19 and/or amendments of the international application under Article 34 are received by the International Preliminary Examining Authority before it has begun to draw up a written opinion or the international preliminary examination report, as so amended.					
Language for the purposes of international preliminary examination:					
which is the language in which the international application was filed.					
which is the language of a translation furnished for the purposes of international search.					
which is the language of publication of the international application.					
which is the language of the translation (to be) furnished for the purposes of international preliminary examination.					
Box No. V ELECTION OF STATES					
The filing of this demand constitutes the election of all Contracting States which are designated and are bound by Chapter II of the					
PCT.					

	Sheet No 4		International application No. PCT/JP2004/005014			
Box No. VI CHECK LIST						
The demand is accompanied by the following elements, in the language referred to in Box No. IV, for the purposes of international preliminary examination:  1. translation of international application : sheets				onal Preliminary uthority use only not received		
*	•	35 sheets		$\overline{\Box}$		
	•	33 5400	اليا			
copy (or, where required, translation) of amendments under Article 19	:	sheets				
<ol> <li>copy (or, where required, translation) of statement under Article 19</li> </ol>	:	sheets				
5. letter	:	sheets				
6. other (specify)	:	sheets				
The demand is also accompanied by the item(s) marked below:  1.  fee calculation sheet  5.  statement explaining lack of signature						
2. original separate power of attorney		6. sequence listing	g in computer readal	ble form		
3. original general power of attorney		7. tables in composequence listin	uter readable form re	elated to a		
4. copy of general power of attorney; reference number, if any:		8. other (specify):	· .	·		
Kubota Yoshitaka KUBOTA, Yoshitaka						
For Internal	ional Preliminar	Examining Authority us	e only			
For International Preliminary Examining Authority use only  1. Date of actual receipt of DEMAND:						
2. Adjusted date of receipt of demand due to CORRECTIONS under Rule 60.1(b):						
3. The date of receipt of the demand is expiration of 19 months from the price item 4 or 5, below, does not apply.	rity date and	expiration item 7 or 8	of the time limit under, below, does not ap			
The applicant has been informed.  4. The date of receipt of the demand is William to f 19 months from the priority data by virtue of Rule 80.5.  5. Although the date of receipt of the demension of 19 months from the priority delay in arrival is EXCUSED pursuant.	THIN the time te as extended and is after the ority date, the	<ul> <li>7.  The date of receipt of the demand is WITHIN the time limit under Rule 54bis.1(a) as extended by virtue of Rule 80.5.</li> <li>8.  Although the date of receipt of the demand is after the expiration of the time limit under Rule 54bis.1(a), the delay in arrival is EXCUSED pursuant to Rule 82.</li> </ul>				
For International Bureau use only						

Demand received from IPEA on:

# **PCT**

## FEE CALCULATION SHEET

## Annex to the Demand

		For International Preliminary Examining Author	rity use only
International application No. PCT/JP2	2004/005014 ~		
Applicant's or agent's file reference DCJ1	31PCT	Date stamp of the IPEA	
Applicant DOW CORNING ASIA LT	D.		
CALCULATION OF PRESCRIB	ed fees	-	
Preliminary examination fee		EUR 1530 P	
2. Handling fee (Applicants from entitled to a reduction of 75% Where the applicant is (or al entitled, the amount to be enter handling fee.)	of the handling fee. I applicants are) so ed at H is 25% of the	EUR 129 H	
Total of prescribed fees     Add the amounts entered at P ar     and enter total in the TOTAL be	nd H	EUR 1659	
MODE OF PAYMENT			
authorization to charge depos account with the IPEA (see be	it cash elow)	stamps	
cheque    X   postal money order	coupons		
bank draft	other (spe	necify):	
	CAD CRENT DEPOSIT	ACCOUNT	
AUTHORIZATION TO CHARGE (This mode of payment may not be ave	rilable at all IPEAs)	IPEA/	
Authorization to charge the tota	I fees indicated above.	Deposit Account No.:	
(This check-box may be marked of deposit accounts of the IPEA so pender charge any deficiency or credit at total fees indicated above.	ermit) Authorization to	Date:  Name:  Signature:	·

#### **AMENDMENT**

To: Examiner of the European Patent Office

1. Identification of the International Application

PCT/JP2004/005014

2. Applicant

Name: DOW CORNING ASIA LTD.

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3. Agent

Name: KUBOTA Yoshitaka Signature:

Address: c/o Asa International Patent Firm Chiba Office 43-27, Satsukigaoka 1-chome, Hanamigawa-ku Chiba-shi, Chiba 2620014 Japan

- 4. Item to be Amended: DESCRIPTION and CLAIMS
- 5. Subject Matter of Amendment
  - (1) The words "Curable Organopolysiloxane Composition for Optical Transmission Components, Optical Transmission Components, and Fabrication Process Thereof" which appear on page 1, lines 3, 4 of the description should be amended as "CURABLE ORGANOPOLYSILOXANE RESIN COMPOSITION FOR OPTICAL WAVEGUIDES, OPTICAL WAVEGUIDE, AND **FABRICATION** PROCESS THEREOF".
  - (2) The sentence "The present invention relates to a curable organopolysiloxane resin composition for optical transmission components,

especially for optical transmission components serving as optical communication elements, relates to optical transmission components, represented by optical waveguides, comprising a hydrosilation-cured product of an organopolysiloxane resin, and relates to a process for fabricating optical transmission components." which appears on page 1, line 7 to 11 of the description should be amended as "The present invention relates to a curable organopolysiloxane resin composition for optical waveguides, especially for optical waveguides serving as optical communication elements, relates to an optical waveguide comprising a hydrosilation-cured product of an organopolysiloxane resin, and relates to a process for fabricating an optical waveguide."

- (3) The words "transmission components" which appear on the following of the description should be amended as "waveguides".

  lines 5, 33, 34 of page 3; line 11 of page 4; lines 35, 38 of page 5; lines 2, 4, 9, 13, 16 of page 6; lines 15, 16 of page 7; line 27 of page 11; lines 26, 29, 30, 31, 32 of page 16; lines 3, 7, 8, 28, 29, 33, 34 of page 17; lines 3, 5, 15, 17, 20, 29 of page 18; lines 26, 27 of page 19; lines 1, 8, 9 of page 20; line 27, 29 of page 22; line 19 of page 23
- (4) The words "transmission component" which appear on the following of the description should be amended as "waveguide". lines 12, 21 of page 3; lines 25, 37 of page 4; lines 12, 16, 20, 25, 30, 34, 37 of page 5; lines 8, 12 of page 6; line 3 of page 10; line 34 of page 14; lines 1, 12, 24 of page 16; lines 1, 4 of page 17
- (5) The words "transmissions component" which appear on page 4, line 8 of the description should be amended as "waveguides".
- (6) The words "The optical transmission component according to [4], wherein the optical transmission component is an optical waveguide." which appear on page 5, lines 8, 9 of the description should be deleted.
- (7) The words "The optical transmission component according to [5], wherein the optical transmission component is an optical waveguide." which appear on page 5, lines 11, 12 of the description should be deleted.
- (8) The words "transmission component consisting of a core and cladding, such as an optical waveguide" which appears on page 8, line 19 should be amended as "waveguide consisting of a core and a cladding".
- (9) The words "an optical transmission component such as" which appear on page 10, lines 19, 20 of the description should be deleted.
- (10) The words "Optical transmission components, such as optical waveguides" which appear on page 10, line 20 and page 15, line 8 of the description should be amended as "optical waveguides".

- (11) The sentences "The optical transmission component of the present invention can be used both as a passive optical transmission component or an active optical transmission component. Its possible uses are exemplified by non-branching waveguides, branching waveguides, optical splitters, optical couplers, optical adhesives, and other passive optical transmission components; waveguide-type optical switches, waveguide-type optical modulator, optical attenuators, optical amplifiers, and other active optical transmission components. In the optical transmission component of the present invention comprising a cured product of organopolysiloxane resin, the optical transmission component may consist of the cured product of organopolysiloxane resin alone, or it may be covered with other material or component, held between other components or members, or inserted into other components or devices." which appear on page 16, lines 12 to 22 of the description should be amended as "The optical waveguide of the present invention can be used both as a passive optical waveguide and an active optical waveguide. Its possible uses are exemplified non-branching waveguides, branching waveguides, optical splitters, optical couplers, and waveguide-type optical switches, waveguide-type optical modulators, optical attenuators, and optical amplifiers. In the optical waveguide of the present invention comprising a cured product of organopolysiloxane resin, the optical waveguide may consist of the cured product of organopolysiloxane resin alone, or it may be covered with other material or component, held between other components or members, or inserted into other components or devices.".
- (12) The words "optical transmission components such as optical waveguides" which appear on page 19, line 32 of the description should be amended as "an optical waveguide".
- (13) The words "Optical transmissions components" which appear on page 19, line 34 of the description should be amended as "An optical waveguide".
- (14) The words "optical transmission components, such as optical waveguides" which appear on page 20, line 15 of the description should be amended as "an optical waveguide".
- (15) The sentences " The curable organopolysiloxane resin composition for optical transmission components of the present invention is useful for fabricating various optical transmission components and is suitable for use as materials for optical communications and optical integrated circuits in the near infrared region, the optical transmission components of the present invention is useful as passive optical transmission components

exemplified by non-branching waveguides, branching waveguides, optical splitters, optical couplers, optical adhesives, and active optical transmission components exemplified by waveguide-type optical switches, waveguide-type optical modulator, optical attenuators, and optical amplifiers, and the process for fabricating optical transmission components of the present invention is useful for fabricating various optical transmission components with lower process cost."which appear on page 27, line 30 to page 28, line 28 of the description should be amended as "The curable organopolysiloxane resin composition for optical waveguides of the present invention is useful for fabricating various optical waveguides and is suitable for use as materials for optical communications and optical integrated circuits in the near infrared region, the optical waveguide of the present invention is useful as passive optical exemplified by non-branching waveguides, waveguides, optical splitters, and optical couplers, and active optical waveguide exemplified by waveguide-type optical waveguide-type optical modulator, optical attenuators, and optical amplifiers, and the process for fabricating an optical waveguide of the present invention is useful for fabricating various optical waveguides with lower process cost.".

- (16) The words "1300~1660-nm" which appear on page 1, line 25 of the description should be amended as "1300 to 1660 nm".
- (17) The mark "~" which appears on the following of the description should be amended as "to".
  - line 25 of page 1; line 34 of page 2; line 38 of page 3; lines 1, 29, 30 of page 4; lines 36, 39 of page 5; lines 3, 10, 17 of page 6; lines 25, 26 of page 7; line 6 of page 8; lines 22, 38 of page 11; line 39 of page 12; line 9 of page 13; lines 6, 28, 38, 39 of page 14; line 35 of page 17; line 36 of page 18; line 30 of page 20; line 1, 14 of page 23
- (18) The mark "]" which appears on page 3, line 4 of the description should be deleted.
- (19) Mark ":" should be added on page 4, lines 13, 26, 33.
- (20) The words "[4]~[11]" which appear on page 5, line 30 should be amended as "[4], [5], [8] to [11]".
- (21) The word "transmissivity" which appears on page 8, line 15; page 17, line 1; page 18, line 15; page 23, Table 1 of the description should be amended as "transmittance".
- (22) The mark ";" which appears on page 9, line 13 should be deleted.
- (23) The word "dimethylpropynoxy" which appears on page 14, line 24 of

- the description should be amended as "dimethylpropinoxy".
- (24) The mark ", " which appears on page 14, line 22 of the description should be amended as ",".
- (25) The words "removing the cured product from the mold" which appear on page 18, line 19 of the description should be amended as "the molding is removed from the mold".
- (26) The words "region. and" which appear on page 20, line 5 of the description should be amended as "region and".
- (27) The word "Phenyl" which appears on page 20, line 39 of the description should be amended as "phenyl".
- (28) The word "propynyloxy" which appears on page 26, lines 9, 17 of the description should be amended as "propinoxy".
- (29) The words "transmission components" which appear on the following of the claims should be amended as "waveguides".
  - lines 3, 17 of page 29; lines 22, 30, 35, 38 of page 31
- (30) The words "transmission component" which appear on the following of the claims should be amended as "waveguide".
  - lines 21, 34 of page 29; lines 9, 26, 31, 36 of page 30; lines 4, 10, 13, 17, 29, 34 of page 31
- (31) Claim 6 and claim 7 should be cancelled.
- (32) The words "claim 6" which appear on page 30, line 26 of the claims should be amended as "claim 4".
- (33) The words "claim 7" which appear on page 30, line 31 of the claims should be amended as "claim 5".
- (34) The words "claim  $4 \sim$  claim 11" which appear on page 31, line 10 of the claims should be amended as "claims 4, 5, 8 to 11".
- (35) The mark "~" which appears on the followings of the claims should be amended as "to".
  - lines 8, 9, 38 of page 29; line 1 of page 30; lines 10, 15, 19, 23, 31 of page 31;
- (36) The numbers "[1]~[3]" which appear on page 31, line 36 and page 31, line 39 of the claims should be amended as "claim 1 to claim 3".

#### 6. List of Attached Documents

(1) Replacement sheet of pages 1, 2, 3, 4, 5, 6, 7, 8, 10, 11,12,13,14,15,16,17,18,19, 20, 22, 23, 26, 27, 28, 29, 30, 31, 32

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#### **DESCRIPTION**

CURABLE ORGANOPOLYSILOXANE RESIN COMPOSITION FOR OPTICAL WAVEGUIDES, OPTICAL WAVEGUIDE, AND FABRICATION PROCESS THEREOF

#### Technical Field

[0001] The present invention relates to a curable organopolysiloxane resin composition for optical waveguides, especially for optical waveguides serving as optical communication elements, relates to an optical waveguide comprising a hydrosilation-cured product of an organopolysiloxane resin, and relates to a process for fabricating an optical waveguide.

## Background of the Invention

[0002] Quartz and glass are used not only as optical fiber materials, but also, being high-reliability materials, as materials for optical communication elements as well. However, such inorganic materials require high-temperature treatment and have inferior productivity characteristics, which creates demand for organic materials for optical communication elements possessing sufficient processability and durability. The most reliable materials are polyimides, which are widely used as materials for electronic components.

- [0003] On the other hand, due to their optical transparency, electrical insulation properties, photostability, thermal stability, etc., organopolysiloxane-based materials have also attracted considerable attention in the field of optoelectronics. Among the physical properties required for optical communication element materials, particular importance is emphasized about the optical characteristics, such as absence of absorption at the 1300 to 1660 nm telecommunication wavelength range and absence of birefringence due to polymer chain orientation, as well as high moisture resistance, low water pick-up, and heat resistance during device assembly. Improvement in the above-described characteristics is in progress focusing primarily on polyimides and organopolysiloxane-based materials.
- [0004] In Japanese Patent Application Publication No. Sho 63-217306, a silicone rubber was offered as a cured product of organopolysiloxane for use in optical waveguides, and an easily deformable organopolysiloxane elastomer was offered as a cured product of organopolysiloxane for use in optical waveguides in Japanese Patent Application Publication No. Hei 1-131505. A liquid addition-curable silicone rubber containing a silica filler has been offered for use as an optical transmission material in Japanese Patent Application Publication No. Hei 1-143605. However, silicone rubbers and

organopolysiloxane elastomers, in view of their molecular structure, exhibit considerable changes in their refractive indices and other optical characteristics following changes in ambient temperature, in other words, they have problems in terms of temperature change stability and heat resistance.

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- [0005] Japanese Patent Application Publication No. Hei 3-43423 offers an organopolysiloxane resin (an organopolysiloxane resin comprising monoorganosiloxane units and diorganosiloxane units) obtained by hydrolytic co-condensation of a diorganodichlorosilane and an organotrichlorosilane having deuterated alkyl or halogenated alkyl groups, as well as an organopolysilsesquioxane obtained by hydrolytic condensation of an organotrichlorosilane having deuterated alkyl or halogenated alkyl groups. Taking into consideration the high optical transmissivity of organopolysiloxane, Japanese Patent Application Publication No. Hei 4-157402 provides an organopolysiloxane resin having hydrocarbon groups without deuteron substitution or halogen substitution. Japanese Patent Application Publication No. Hei 9-124793 offers an epoxy-containing phenylpolysiloxane resin obtained by the hydrolytic co-condensation of phenyltrichlorosilane and diphenyldichlorosilane in the presence of an epoxy-containing alcohol.
- [0006] However, such organopolysiloxane resins, although possessing superior optical characteristics at normal temperatures, are of the types that cure by dehydration condensation of silanol groups, and, as a result, do not sufficiently cure without heating at an elevated temperature for an extended period of time, and when an optical transmission component, such as an optical waveguide, is exposed to elevated temperatures (e.g., about 260°C) during fabrication of an optical communication device, etc., further dehydration condensation takes place, resulting in changes in optical characteristics.
  - [0007] In Japanese Patent Application Publication No. Hei 9-124793, UV curing is implemented by adding a photocuring catalyst to an epoxy-containing phenylpolysiloxane resin, however, such addition leads to problems including increased absorption of communication light and a tendency towards scattering.
  - [0008] Incidentally, an optical waveguide is made up of two types of materials with different refractive indices called a core and a cladding. The difference in their refractive indices depends on the design of the optical waveguide, but it is said that the refractive index of the core has to be about 0.1% to 5% higher than the refractive index of the cladding in order for light to propagate through the core. In case of a dehydration condensation-curable organopolysiloxane resin, the refractive index difference is regulated by the amount of introduced fluorinated hydrocarbon groups which contribute to a reduction in the refractive index. For instance, Japanese Patent Application Publication No. 2000-230052 offers an organopolysilsesquioxane having fluorinated hydrocarbon groups, but the problem with this approach, however, consists in the

increased cost of the material due to the introduction of the fluorinated hydrocarbon groups.

[0009] It is an object of the present invention to provide a curable organopolysiloxane resin composition for optical waveguides that has low temperature dependence of the refractive index, high heat resistance, high transparency at the telecommunication wavelength region without introducing deuterated alkyl groups and fluorinated hydrocarbon groups, possesses elasticity and hardness that makes it difficult to deform, and permits easy adjustment of the refractive index difference during the preparation of the core material and cladding material.

[0010] Furthermore, its object is also to provide an optical waveguide comprising a cured product of organopolysiloxane resin that has low temperature dependence of the refractive index, high heat resistance, high transparency at the telecommunication wavelength region without introducing deuterated alkyl groups and fluorinated hydrocarbon groups, possesses elasticity and hardness that makes it difficult to deform, and permits easy adjustment of the refractive index difference during the preparation of the core material and cladding material.

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[0011] Furthermore, it is an object of the invention to provide a simple and economical process for fabricating an optical waveguide comprising a cured product of organopolysiloxane resin that that has low temperature dependence of the refractive index, high heat resistance, high transparency at the telecommunication wavelength region without introducing deuterated alkyl groups and fluorinated hydrocarbon groups, possesses elasticity and hardness that makes it difficult to deform, and permits easy adjustment of the refractive index difference during the preparation of the core material and cladding material.

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#### Summary of the Invention

[0012] The inventors arrived at the present invention as a result of in-depth investigations aimed at resolving the above-mentioned problems. Namely, the present invention relates to the following;

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[0013] [1] A curable organopolysiloxane resin composition for optical waveguides comprising(A) an organopolysiloxane resin, which is represented by the average unit formula (1):

$$(R^{1}_{3}SiO_{1/2})_{a}(R^{2}_{2}SiO_{2/2})_{b}(R^{3}SiO_{3/2})_{c}(SiO_{4/2})_{d}$$
 (1)

(wherein R<sup>1</sup>, R<sup>2</sup>, and R<sup>3</sup> stand for one, two, or more kinds of monovalent hydrocarbon groups selected from monovalent aliphatic hydrocarbon groups having 1 to 6 carbon

atoms and monovalent aromatic hydrocarbon groups having 6 to 10 carbon atoms,  $0 < a \le 0.5$ ,  $0 \le b < 0.2$ ,  $0.3 \le c < 1$ ,  $0 \le d \le 0.4$ ,  $0 \le (b+d)/(a+c) \le 0.25$ , and a+b+c+d=1) and has three or more monovalent unsaturated aliphatic hydrocarbon groups per molecule, with not less than 10 mol% of the monovalent hydrocarbon groups being monovalent aromatic hydrocarbon groups. (B) an organosilicon compound having two or more silicon-bonded

- hydrocarbon groups, (B) an organosilicon compound having two or more silicon-bonded hydrogen atoms per molecule, with not less than 5 mol% of all the silicon-bonded monovalent substituent groups being monovalent aromatic hydrocarbon groups, and (C) a hydrosilation catalyst.
- [2] The curable organopolysiloxane resin composition for optical waveguides according to [1], wherein the viscosity of the composition is not more than 1×10<sup>7</sup> mPa·s at 25°C.
  - [3] A curable organopolysiloxane resin composition for optical waveguides comprising (A) an organopolysiloxane resin, which is represented by an average unit formula (1):  $(R^{1}_{3}SiO_{1/2})_{a}(R^{2}_{2}SiO_{2/2})_{b}(R^{3}SiO_{3/2})_{c}(SiO_{4/2})_{d}$  (1)
- (wherein R<sup>1</sup>, R<sup>2</sup>, R<sup>3</sup>, a, b, c, d, (b+d)/(a+c), and a+b+c+d are the same as above) and has three or more monovalent unsaturated aliphatic hydrocarbon groups per molecule, with not less than 10 mol% of the monovalent hydrocarbon groups being monovalent aromatic hydrocarbon groups, (B) an organosilicon compound having two or more silicon-bonded hydrogen atoms per molecule, with not less than 5 mol% of all the silicon-bonded monovalent substituent groups being monovalent aromatic hydrocarbon groups, (C) a hydrosilation catalyst, and (D) (d1) a solvent or (d2) a hydrosilation-reactive organosiloxane-based diluent.

## 25 [0014]

- [4] An optical waveguide comprising a hydrosilation-cured product of (A) an organopolysiloxane resin, which is represented by an average unit formula (1): (R<sup>1</sup><sub>3</sub>SiO<sub>1/2</sub>)<sub>a</sub>(R<sup>2</sup><sub>2</sub>SiO<sub>2/2</sub>)<sub>b</sub>(R<sup>3</sup>SiO<sub>3/2</sub>)<sub>c</sub>(SiO<sub>4/2</sub>)<sub>d</sub> (1)
- (wherein R<sup>1</sup>, R<sup>2</sup>, and R<sup>3</sup> stand for one, two, or more kinds of monovalent hydrocarbon groups selected from monovalent aliphatic hydrocarbon groups having 1 to 6 carbon atoms and monovalent aromatic hydrocarbon groups having 6 to 10 carbon atoms, 0<a≤0.5, 0≤b<0.2, 0.3≤c<1, 0≤d≤0.4, 0≤(b+d)/(a+c)≤0.25, and a+b+c+d=1) and has three or more monovalent unsaturated aliphatic hydrocarbon groups per molecule, with not less than 10 mol% of the monovalent hydrocarbon groups being monovalent aromatic hydrocarbon groups and (R) an organosilison compound having two or more silison.
- hydrocarbon groups, and (B) an organosilicon compound having two or more siliconbonded hydrogen atoms per molecule, with not less than 5 mol% of all the silicon-bonded monovalent substituent groups being monovalent aromatic hydrocarbon groups.
  - [5] An optical waveguide comprising a hydrosilation-cured product of (A) an organopolysiloxane resin, which is represented by an average unit formula (1):
- (R<sup>1</sup><sub>3</sub>SiO<sub>1/2</sub>)<sub>a</sub>(R<sup>2</sup><sub>2</sub>SiO<sub>2/2</sub>)<sub>b</sub>(R<sup>3</sup>SiO<sub>3/2</sub>)<sub>c</sub>(SiO<sub>4/2</sub>)<sub>d</sub> (1) (wherein R<sup>1</sup>, R<sup>2</sup>, R<sup>3</sup>, a, b, c, d, (b+d)/(a+c), and a+b+c+d are the as described above) and has three or more monovalent unsaturated aliphatic hydrocarbon groups per molecule,

with not less than 10 mol% of the monovalent hydrocarbon groups being monovalent aromatic hydrocarbon groups, (B) an organosilicon compound having two or more silicon-bonded hydrogen atoms per molecule, with not less than 5 mol% of all the silicon-bonded monovalent substituent groups being monovalent aromatic hydrocarbon groups, and (d2) a hydrosilation-reactive organosiloxane-based diluent.

[6]

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[7]

- 10 [8] The optical waveguide according to [4], wherein both the cladding and the core of the optical waveguide consist of a hydrosilation-cured product of component (A) and component (B), with the refractive index of the core being at least 0.1% higher than the refractive index of the cladding.
- [9] The optical waveguide according to [5], wherein both the cladding and the core of the optical waveguide consist of a hydrosilation-cured product of component (A), component (B), and component (d2), with the refractive index of the core being at least 0.1% higher than the refractive index of the cladding.
  - [10] The optical waveguide according to [8], wherein the refractive index difference is regulated by making the total content of monovalent aromatic hydrocarbon groups in component (A) and component (B) used for the core higher than the total content of monovalent aromatic hydrocarbon groups in component (A) and component (B) used for the cladding.
  - [11] The optical waveguide according to [9], wherein the refractive index difference is regulated by making the total content of monovalent aromatic hydrocarbon groups in component (A), component (B), and component (d2) used for the core higher than the total content of monovalent aromatic hydrocarbon groups in component (A), component (B), and component (d2) used for the cladding.
  - [12] The optical waveguide according to any of [4], [5], [8] to [11], which has a film-like shape.

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[0015]

- [13] A process for fabricating an optical waveguide, wherein the curable organopolysiloxane resin composition for optical waveguides according to any of [1] to [3] is cured by heating.
- [14] A process for fabricating an optical waveguide, wherein the curable organopolysiloxane resin composition for optical waveguides according to any of [1] to [3] is applied to a substrate and cured by heating.
  - [15] A process for fabricating a slab optical waveguide, in which a curable organopolysiloxane resin composition for optical waveguides (1) according to any of [1]
- to [3] is applied to a substrate and cured by heating, a curable organopolysiloxane resin composition for optical waveguides (2), whose cured product has a refractive index at least 0.1% higher than that of the above-mentioned composition (1), is applied to the

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cured product thereof and cured by heating, and then the aforementioned composition (1) is applied to the cured product thereof and cured by heating.

- [16] A process for fabricating an optical waveguide, wherein the curable organopolysiloxane resin composition for optical waveguides according to any of [1] to [3] is cast into a mold having a desired inner surface shape and cured by heating.
- [17] A process for fabricating an optical waveguide, wherein ① a curable organopolysiloxane resin composition for optical waveguides (3) according to any of [1] to [3] is cast into a mold having on its inner surface protrusions corresponding to the core and cured by heating, ② the molding is removed from the mold, ③ a curable organopolysiloxane resin composition for optical waveguides (4) according to any of [1] to [3], whose cured product has a refractive index at least 0.1% higher than that of the aforementioned composition (3), is cast into the hollow portion of the cured product removed from the mold and cured by heating, whereupon ④ the aforementioned composition (4) and the cured product of the aforementioned composition (5) and cured by heating.

## Brief Description of the Drawings

- [0016] FIG. 1 is a cross-sectional view of the slab type optical waveguide of Example 3.
- FIG. 2 is a cross-sectional view of the mold used in the process of fabrication of the channel type optical waveguide of Example 4.
  - FIG. 3 is a cross-sectional view of the mold used in the process of fabrication of the channel type optical waveguide of Example 4.
- FIG. 4 is a cross-sectional view of the mold used in the process of fabrication of the channel type optical waveguide of Example 4.
  - FIG. 5 is a cross-sectional view of the mold used in the process of fabrication of the channel type optical waveguide of Example 4.
  - FIG. 6 is a cross-sectional view of the mold used in the process of fabrication of the channel type optical waveguide of Example 4.
- FIG. 7 is a cross-sectional view of the mold used in the process of fabrication of the channel type optical waveguide of Example 4.
  - FIG. 8 is a cross-sectional view of the channel type optical waveguide of Example 4.
  - [0017] The reference numerals in the drawings are as follows;
- A is Slab type optical waveguide,
  - B is Channel type optical waveguide,
  - 1 is Cured product of composition 5,
  - 2 is Cured product of composition 3,
  - 3 is First mold,
- 40 4 is Protrusion.
  - 5 is Composition 4 (does not contain toluene),
  - 6 is Second mold,

7 is Cured product of composition 4 (does not contain toluene), 8 is Hollow portion,

9 is Cured product of composition 3 (does not contain toluene), and 10 is Third mold.

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#### Best Mode for Carrying Out the Invention

[0018] In the curable organopolysiloxane resin composition for optical waveguides of the present invention, monovalent unsaturated aliphatic hydrocarbon groups of component (A) and silicon-bonded hydrogen atoms of component (B), or monovalent unsaturated aliphatic hydrocarbon groups of component (A) and component (d2) and silicon-bonded hydrogen atoms of component (B) are crosslinked and cured by a hydrosilation reaction under the action of component (C).

[0019] As component (A) is represented by an average unit formula (1)

(R¹₃SiO₁/₂)a(R²₂SiO₂/₂)b(R³SiO₃/₂)c(SiO₄/₂)d (1)

(wherein R¹, R², and R³ stand for one, two, or more kinds of monovalent hydrocarbon groups selected from monovalent aliphatic hydrocarbon groups having 1 to 6 carbon atoms and monovalent aromatic hydrocarbon groups having 6 to 10 carbon atoms, 0<a≤0.5, 0≤b<0.2, 0.3≤c<1, 0≤d≤0.4, 0≤(b+d)/(a+c)≤0.25, and a+b+c+d=1), a and c are never zero, but b and d may be equal to zero, which is why unit R¹₃SiO₁/₂ and unit R³SiO₃/₂are essential units while unit R²₂SiO₂/₂ and unit SiO₄/₂ are optional units.

[0020] Therefore, there can be organopolysiloxane resins comprising the following units.

25  $(R_3^1SiO_{1/2})_a(R_3^3SiO_{3/2})_c$ ,  $(R_3^1SiO_{1/2})_a(R_2^2SiO_{2/2})_b(R_3^3SiO_{3/2})_c$ ,  $(R_3^1SiO_{1/2})_a(R_3^3SiO_{3/2})_c(SiO_{4/2})_d$ , and  $(R_3^1SiO_{1/2})_a(R_2^2SiO_{2/2})_b(R_3^3SiO_{3/2})_c(SiO_{4/2})_d$ . However, too many units  $R_3^1SiO_{1/2}$  bring about a decrease in the molecular weight, which is why  $0 \le a \le 0.5$ , and preferably,  $0 \le a \le 0.3$ . Too few units  $R_3^3SiO_{3/2}$  bring about a decrease in the degree of branching, which is why  $0.3 \le c \le 1$ , and preferably,  $0.5 \le c \le 1$ .

While introducing units R<sup>2</sup><sub>2</sub>SiO<sub>2/2</sub> into the organopolysiloxane resin generally results in an increase in the elasticity of the resin, this can cause a decrease in its thermal deformation temperature and is an important factor affecting shape changes. For this reason, 0≤b<0.2, and preferably 0≤b<0.1. On the other hand, introduction of SiO<sub>4/2</sub> units results in a significant increase in the hardness of the resin, and the resin tends to become brittle. Thus, 0≤d≤0.4 and 0≤(b+d)/(a+c)≤0.25.

[0021] Monovalent aliphatic hydrocarbon groups having 1 to 6 silicon-bonded carbon atoms include methyl, ethyl, propyl, butyl, hexyl, and other monovalent saturated aliphatic hydrocarbon groups, as well as vinyl, allyl, hexenyl, and other monovalent unsaturated aliphatic hydrocarbon groups. Silicon-bonded monovalent aromatic hydrocarbon groups are exemplified by phenyl, tolyl, xylyl, and naphthyl. Said

monovalent aliphatic hydrocarbon groups are preferably methyl, said monovalent unsaturated aliphatic hydrocarbon groups preferably vinyl, and said monovalent aromatic hydrocarbon groups are preferably phenyl.

In the organopolysiloxane resin, 10 mol% or more of its silicon-bonded monovalent hydrocarbon groups have to be monovalent aromatic hydrocarbon groups. Less than 10 mol% brings about a decrease in transmittance in the communication wavelength region of the cured product of the organopolysiloxane resin.

[0022] When an organopolysiloxane resin is used for the core section in an optical waveguide consisting of a core and cladding, monovalent aromatic hydrocarbon groups preferably comprise 20 mol% or more. The refractive index, which is an important optical characteristic, is adjusted by changing the type of the monovalent hydrocarbon groups. Using substituent groups consisting mainly of methyl and other monovalent aliphatic hydrocarbon groups tends to make the refractive index less than 1.5, and using substituent groups consisting mainly of phenyl and other monovalent aromatic hydrocarbon groups tends to make the refractive index higher than 1.5.

[0023] Organopolysiloxane resin consisting of (PhSiO<sub>3/2</sub>) and (Me<sub>2</sub>ViSiO<sub>1/2</sub>) units, organopolysiloxane resin consisting of (PhSiO<sub>3/2</sub>), (Me<sub>2</sub>SiO<sub>2/2</sub>), and (Me<sub>2</sub>ViSiO<sub>1/2</sub>) units, organopolysiloxane resin consisting of (PhSiO<sub>3/2</sub>), (MeSiO<sub>3/2</sub>), and (Me<sub>2</sub>ViSiO<sub>1/2</sub>) units, **20** organopolysiloxane resin consisting of (PhSiO<sub>3/2</sub>), (MeSiO<sub>3/2</sub>), (Me<sub>2</sub>ViSiO<sub>1/2</sub>), and (SiO<sub>4/2</sub>) units, organopolysiloxane resin consisting of (PhSiO<sub>3/2</sub>), (Me<sub>2</sub>ViSiO<sub>1/2</sub>), and (SiO<sub>4/2</sub>) units, organopolysiloxane resin consisting of (PhSiO<sub>3/2</sub>), (Ph<sub>2</sub>SiO<sub>2/2</sub>), and  $(Me_2ViSiO_{1/2})$  units, organopolysiloxane resin consisting of  $(MeSiO_{3/2})$ ,  $(Ph_2SiO_{2/2})$ , and 25  $(Me_2ViSiO_{1/2})$  units, organopolysiloxane resin consisting of  $(PhSiO_{3/2})$  and (MePhViSiO<sub>1/2</sub>) units, organopolysiloxane resin consisting of (PhSiO<sub>3/2</sub>), (MeSiO<sub>3/2</sub>), and (MePhViSiO<sub>1/2</sub>) units, and organopolysiloxane resin consisting of (PhSiO<sub>3/2</sub>), (MePhViSiO<sub>1/2</sub>), and (SiO<sub>4/2</sub>) units [where Me stands for methyl, Ph for phenyl, and Vi for vinyl] are suggested as specific examples of the organopolysiloxane resins. Same 30 below]. Two or more kinds of such organopolysiloxane resins can be used in combination. These organopolysiloxane resins usually have reticular and threedimensional structures.

[0024] Processes used in the preparation of such organopolysiloxane resins are well known (for example, see Kunio Itoh's "Silicone Handbook", (published by Nikkan Kogyo Shinbunsha, 1990), pp. 468~470, or M. Wada, "Newest Silicone Technology: Research and Applications" (published by CMC, 1986), pp. 80~81). For instance, they can be easily prepared by hydrolytic co-condensation of corresponding organochlorosilanes or organoalkoxysilanes in an organic solvent, or by hydrolytic co-condensation of corresponding organosiloxane oligomers and organoalkoxysilanes in an organic solvent in the presence of a strong acid catalyst.

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[0029] The monovalent aromatic hydrocarbon groups are exemplified by phenyl, tolyl, xylyl, and naphthyl, with phenyl being preferable. Organic groups other than the monovalent aromatic hydrocarbon groups are preferably the above-mentioned monovalent saturated aliphatic hydrocarbon groups, with methyl being preferable. In addition, when an optical waveguide is fabricated by curing component (A) and the present component via a hydrosilation reaction, high volatility leads to insufficient curing, which is why low volatility compounds are more preferable. Specifically, compounds with a boiling point of less than 200°C under normal pressure are undesirable.

10 [0030] Diphenylsilane, 1,2-bis(dimethylsilyl)benzene, 1,4-bis(dimethylsilyl)benzene and other organosilanes having two silicon-bonded hydrogen atoms; phenyltris(dimethylsiloxy)silane, tris(methylphenylsiloxy)silane, tetra(methylphenylsiloxy)silane and other organosiloxane oligomers having three or four silicon-bonded hydrogen atoms;

[0031] Organopolysiloxane resin consisting of (PhSiO<sub>3/2</sub>) and (Me<sub>2</sub>HSiO<sub>1/2</sub>) units, organopolysiloxane resin or branched organosiloxane oligomer consisting of (PhSiO<sub>3/2</sub>), (Me<sub>2</sub>SiO<sub>2/2</sub>), and (Me<sub>2</sub>HSiO<sub>1/2</sub>) units, organopolysiloxane resin or branched organosiloxane oligomer consisting of (PhSiO<sub>3/2</sub>), (MeSiO<sub>3/2</sub>), and (MeHSiO<sub>1/2</sub>) units, organopolysiloxane resin or branched organosiloxane oligomer consisting of (PhSiO<sub>3/2</sub>) and (MeHSiO<sub>2/2</sub>) units, organopolysiloxane resin or branched organosiloxane oligomer consisting of (Me<sub>2</sub>HSiO<sub>1/2</sub>), (MePh<sub>2</sub>SiO<sub>1/2</sub>) and (SiO<sub>4/2</sub>) units; linear organopolysiloxane or organosiloxane oligomer consisting of (MePhSiO<sub>2/2</sub>) and (Me<sub>2</sub>HSiO<sub>1/2</sub>) units, linear organopolysiloxane or organosiloxane oligomer consisting of (MePhSiO<sub>2/2</sub>), (MePhSiO<sub>2/2</sub>) and (Me<sub>2</sub>HSiO<sub>1/2</sub>) units, linear organopolysiloxane or organosiloxane oligomer consisting of (MePhSiO<sub>2/2</sub>), (MeHSiO<sub>2/2</sub>) and (Me<sub>3</sub>SiO<sub>1/2</sub>) units, linear organopolysiloxane or organosiloxane oligomer consisting of (MePhSiO<sub>2/2</sub>) and (Me<sub>2</sub>HSiO<sub>1/2</sub>) and (Me<sub>2</sub>HSiO<sub>1/2</sub>)

and  $(Me_3SiO_{1/2})$  units, linear organopolysiloxane or organosiloxane oligomer consisting of  $(MeHSiO_{2/2})$  and  $(MePh_2SiO_{1/2})$  units, and cyclic organopolysiloxane or organosiloxane oligomer consisting only of  $(PhHSiO_{2/2})$  units are suggested as specific examples.

units, linear organopolysiloxane or organosiloxane oligomer consisting of (PhHSiO<sub>2/2</sub>)

[0032] Two or more kinds of these organosilicon compounds can be used in combination. Processes used in the preparation of these organosilicon compounds are publicly known or well-known, and such compounds, for instance, can be produced via a hydrolytic condensation reaction using only organochlorosilanes containing siliconbonded hydrogen atoms, or by means of a hydrolytic co-condensation reaction between organochlorosilanes having silicon-bonded hydrogen atoms and organochlorosilanes having no silicon-bonded hydrogen atoms.

[0033] The hydrosilation catalysts of component (C) are metals of Group VIII of the Periodic Table or their compounds, with platinum and platinum compounds being preferable. They are exemplified by microparticulate platinum, chloroplatinic acid, platinum-diolefin complexes, platinum-diketone complexes, platinum-

divinyltetramethyldisiloxane complexes, and platinum-phosphine complexes. The amount, in which it is added, is in the range of 0.05 to 300 ppm, and, more preferably, in the range of 0.1 to 50 ppm in terms of the weight of the metal relative to the total weight of component (A) and component (B). Below this range, the crosslinking reaction may not proceed to a sufficient extent, while exceeding the range is unnecessary and may cause deterioration in the optical properties due to the residual metal.

[0034] The organopolysiloxane resin composition for optical waveguides of the present invention is obtained by uniformly mixing component (A), component (B) and component (C). When these three components are mixed, the crosslinking reaction takes place even at normal temperature, causing an increase in viscosity and ultimately curing, which is why it is better to either prepare a mixture of component (A) and component (B) and combine component (C) with it at the time of molding, or prepare a mixture of component (A) and component (C) and combine component (B) with it at the time of molding.

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[0035] The proportion, in which component (A) and component (B) are mixed, is such that the mole ratio of silicon-bonded hydrogen atoms in component (B) to unsaturated aliphatic groups in component (A) is preferably 0.2 to 5.0, and even more preferably, 0.7 to 1.5. The proportion, in which component (A) and component (B) are mixed, is preferably varied depending on the intended use of the resultant cured product.

[0036] If the cured product is to be used in the near ultraviolet wavelength region, it is desirable to make sure no unsaturated aliphatic hydrocarbon groups remain after curing by adjusting the amount of silicon-bonded hydrogen atoms such that it slightly exceeds the amount of all the unsaturated aliphatic hydrocarbon groups. The residual silicon-bonded hydrogen atoms have no absorption bands in this region and will not impair optical characteristics.

35 [0037] On the other hand, if the cured product is to be used in the near infrared wavelength region, it is desirable to make sure no silicon-bonded hydrogen atoms remain after curing by adjusting the amount of unsaturated aliphatic hydrocarbon groups such that it slightly exceeds the amount of all the silicon-bonded hydrogen atoms. This is due to the fact that silicon-bonded hydrogen atoms undergo oxidation under the action of the environment when the cured product is used and may turn into silicon-bonded hydroxyl groups that exhibit absorption in this wavelength region, with the hydroxyl groups functioning as sites for adsorption of ambient moisture.

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[0038] While the viscosity of the curable organopolysiloxane resin composition of the present invention varies depending on the molding process used, preferably, it is not more than  $1 \times 10^7$  mPa·s, and even more preferably, not more than  $1 \times 10^6$  mPa·s at 25°C. In case of spin-coating on a substrate at normal temperature, it is preferably not more than  $1 \times 10^4$  mPa·s at 25°C. At a higher viscosity it is difficult to obtain a film of uniform thickness when spin-coating the composition on a substrate. While there are no particular limitations concerning the lower boundary value of viscosity, typically, it is greater than 500 mPa·s at 25°C because it is influenced by the viscosity of component (A) and is naturally higher than the viscosity of component (d2). When the viscosity of component (A) and component (B), and especially that of component (A), is too high and unsuitable for spin coating, the composition is preferably diluted with (d1) a solvent. When the viscosity of component (A) and component (B), and especially that of component (A), is too high and makes the composition unsuitable for casting in a mold, the composition is preferably diluted with (d2) a hydrosilation-reactive organosiloxane-based diluent.

[0039] Solvents with a boiling point of 80 to 200°C are recommended as solvent (d1). They are specifically exemplified by isopropyl alcohol, t-butyl alcohol, methyl ethyl ketone, methyl isobutyl ketone, toluene, xylene, mesitylene, chlorobenzene, ethylene glycol dimethyl ether, ethylene glycol diethyl ether, diethylene glycol dimethyl ether, ethoxy-2-propanol acetate, methoxy-2-propanol acetate, octamethylcyclotetrasiloxane, and hexamethyldisiloxane. Such a solvent (d1) can be used singly or as a combination of two or more solvents.

[0040] While the concentration of solid matter in the composition diluted with solvent depends on the thickness of the coating film to be formed and the viscosity and molecular weight of component (A) and component (B), and especially, on those of component (A), it is desirable that the concentration should be not less than 20 wt%. In addition, the viscosity of the composition diluted with solvent (d1) that is suitable for forming thin
 films of excellent quality using a general-purpose spin coating machine is preferably 10 to 1×10<sup>4</sup> mPa·s at 25°C.

[0041] The hydrosilation-reactive organosiloxane-based diluent (d2) is effective in lowering the viscosity of the composition without significantly impairing the physico-mechanical properties of the cured product of organopolysiloxane resin. It is recommended to use organosiloxane oligomers having at least two monovalent unsaturated aliphatic hydrocarbon groups per molecule, wherein 5 mol% ore more of all the monovalent hydrocarbon groups in the molecule are monovalent aromatic hydrocarbon groups and the number of silicon atoms is not more than 15.

[0042] Their viscosity at 25°C should preferably be not more than 500mPa·s, and even more preferably, not more than 300 mPa·s. Such organosiloxane oligomers have

excellent compatibility with component (B) and component (A) having monovalent aromatic hydrocarbon groups, take part in the crosslinking reaction, and have high transparency in the communication wavelength region of the cured product of organopolysiloxane resin.

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[0043] This component is exemplified by 1,3-divinyldimethyldiphenyldisiloxane, 1,5-divinyltetramethyldiphenyltrisiloxane, phenyltris(dimethylvinylsiloxy)silane, and methylvinylphenylsioxane oligomer consisting of (PhSiO $_{3/2}$ ) and (Me2ViSiO $_{1/2}$ ) units.

10 [0044] Because component (d2) has a negative effect on the storage stability of the composition and causes a decrease in the heat resistance and temperature change stability of the cured product of organopolysiloxane resin when its molecules contain silanol groups and silicon-bonded alkoxy groups, preferably, it should contain only trace amounts, or be completely free of silanol groups and silicon-bonded alkoxy groups. As
15 the amount of component (d2) added to the composition increases, the mechanical strength of the cured product of organopolysiloxane resin decreases to some extent depending on its molecular structure

[0045] For this reason, the amount, in which it is added, should be determined with a view to strike a balance between such physical properties. The recommended amount to be added is such that the weight ratio of [reactive diluent]/[component (A) + component (B)] is in the range of from 5/95 to 80/20, and, even better, the recommended amount is such that the ratio is in the range of from 5/95 to 40/60. The viscosity of the composition diluted with component (d2) is preferably 20 to 1×10<sup>4</sup> mPa·s at 25°C.

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[0046] Because the curable organopolysiloxane resin composition comprising component (A), component (B), component (C) and solvent (d1) is liquid at normal temperature and possesses excellent fluidity, the composition is particularly suitable for spin coating. Because the curable organopolysiloxane resin composition comprising component (A), component (B), component (C) and hydrosilation reactive organosiloxane-based diluent (d2) is liquid at normal temperature and possesses excellent fluidity, the composition is particularly suitable for cast molding.

[0047] When component (A), component (B) and component (C) are mixed, the crosslinking reaction takes place even at normal temperature, causing an increase in viscosity and ultimately curing, which is why a cure retarder is added to the composition, if necessary, in order to prevent an increase in viscosity and curing at normal temperature and facilitate curing under heating. The cure retarders include compounds typically used in hydrosilation-curable compositions, such as, for instance, 3-methyl-1-butyne-3-ol, 3,5-dimethyl-1-hexyne-3-ol, phenylbutynol, and other alkyene alcohols; 3-methyl-3-pentene-1-yne, 3,5-dimethyl-1-hexyne-3-yne and other ene-yne compounds; alkynyl-containing ketones; methyltris(1,1-dimethylpropinoxy)silane, dimethyldi(1,1-

dimethylpropinoxy)silane, and other alkynyl-containing organosilanes; benzotriazole; maleic acid esters, and fumaric acid esters.

[0048] It is recommended that the amount, in which they are added, should be such that the weight ratio of [cure retarder]/[metal in hydrosilation catalyst] is 10 to 10,000. When a large amount of the above-mentioned cure retarder is added, the composition can be a one-package organopolysiloxane resin composition comprising all the components. In addition, so long as the object of the invention is not impaired, it can be combined with adhesion promoters represented by silane coupling agents, and with other additives.

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[0049] The optical waveguide of the present invention comprises a hydrosilation-cured product of (A) an organopolysiloxane resin, which is represented by an average unit formula (1): (R¹3SiO₁/2)a(R²2SiO₂/2)b(R³SiO₃/2)c(SiO₄/2)d (1) (wherein R¹, R², and R³ stand for one, two, or more kinds of monovalent hydrocarbon groups selected from monovalent aliphatic hydrocarbon groups having 1 to 6 carbon atoms and monovalent aromatic hydrocarbon groups having 6 to 10 carbon atoms, 0<a≤0.5, 0≤b<0.2, 0.3≤c<1, 0≤d≤0.4, 0≤(b+d)/(a+c)≤0.25, and a+b+c+d=1) and has three or more monovalent unsaturated aliphatic hydrocarbon groups per molecule, with not less than 10 mol% of the monovalent hydrocarbon groups being monovalent aromatic hydrocarbon groups, and (B) an organosilicon compound having two or more silicon-bonded hydrogen atoms per molecule, with not less than 5 mol% of all the silicon-bonded monovalent substituent groups being monovalent aromatic hydrocarbon groups.

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[0050] Optical waveguides, generally consist of a core having high refractive index and a cladding having a low refractive index. In the organopolysiloxane resin of component (A), both when used for the core and for the cladding, 10 mol% or more of its siliconbonded monovalent hydrocarbon groups have to be monovalent aromatic hydrocarbon groups. In the organosilicon compound of component (B), not less than 5 mol% of all its silicon-bonded monovalent substituent groups are monovalent aromatic hydrocarbon groups. If this requirement regarding their content is not satisfied, it would be detrimental because of the decrease in the transparency of the cured product of organopolysiloxane resin in the communication wavelength region.

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[0051] The refractive index of the cured product of organopolysiloxane resin can be adjusted using the ratio of silicon-bonded monovalent aliphatic hydrocarbon groups (typically, methyl) to monovalent aromatic hydrocarbon groups (typically, phenyl). An increase in the proportion of monovalent aromatic hydrocarbon groups brings about an increase in the refractive index, while increasing the proportion of monovalent aliphatic hydrocarbon groups causes a decrease in the refractive index. Because it is preferable to make the refractive index of the cured product of organopolysiloxane resin used for the cladding, it is preferable to make the content of monovalent aromatic hydrocarbon

groups in the organopolysiloxane resin used for the core higher than that in the organopolysiloxane resin used for the cladding.

[0052] Thus, the refractive index of the cured product used for the core can be made higher than the refractive index of the cured product used for the cladding via a process, in which two kinds of organopolysiloxane resin with different mole ratios of [monovalent aliphatic hydrocarbon group]/[monovalent aromatic hydrocarbon groups] are used separately for the core and for the cladding, and/or a process, in which two kinds of component (B) with different mole ratios of [monovalent aliphatic hydrocarbon groups]/[monovalent aromatic hydrocarbon groups] are used separately for the core and for the cladding. Here, details concerning component (A) and component (B) are as described in the section on the components of the curable organopolysiloxane resin composition.

15 [0053] The optical waveguide comprising the cured product of organopolysiloxane resin of the present invention has a nearly 100% optical transparency if reflection in the visible light range is eliminated. In addition, so long as polarization is determined using a polarizer, no polarization can be observed and birefringence is so small that it can be ignored. Also, because the cured product preserves its original shape and exhibits no appreciable weight changes even when heated to 260°C, it can be said to possess heat resistance that is higher than that of thermoplastic resins used as optical materials, such as fluorinated polymethylmethacrylate resin. Furthermore, the cured product possesses elasticity and hardness to the extent that it does not easily bend and has sufficient self-maintaining strength.

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[0054] The optical waveguide of the present invention can be used both as a passive optical waveguide and an active optical waveguide. Its possible uses are exemplified by non-branching waveguides, branching waveguides, optical splitters, optical couplers, and waveguide-type optical switches, waveguide-type optical modulators, optical attenuators, and optical amplifiers. In the optical waveguide of the present invention comprising a cured product of organopolysiloxane resin, the optical waveguide may consist of the cured product of organopolysiloxane resin alone, or it may be covered with other material or component, held between other components or members, or inserted into other components or devices.

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[0055] The optical waveguide of the present invention comprising a cured product of organopolysiloxane resin is fabricated by heating and curing the above-mentioned curable organopolysiloxane resin composition for optical waveguides. The processes of

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fabrication can be roughly divided into the following two categories. In other words, there is a process for fabricating an optical waveguide, in which the curable organopolysiloxane resin composition for optical waveguides is coated onto a substrate and cured by heating, and a process for fabricating an optical waveguide, in which the curable organopolysiloxane resin composition for optical waveguides is cast and cured by heating in a mold having a desired shape of the inner surface. Injection molding, extrusion molding and other fabrication processes can be also used.

10 [0056] The substrate used in the first fabrication process preferably has a flat surface and is stable to solvents and temperatures used during curing. It is exemplified by silicon wafers, glass, ceramics, and heat-resistant plastics.

[0057] An optical waveguide with a high transmittance in the designated wavelength region is fabricated by coating the above-mentioned curable organopolysiloxane resin composition for optical waveguides onto the substrate and curing it by heating, but, if necessary, a film-like optical waveguide is obtained by peeling the cured product from the substrate. In addition, a slab optical waveguide can be fabricated by ① applying the above-mentioned curable organopolysiloxane resin composition for optical waveguides (1) to a substrate and curing it by heating, ② applying a curable organopolysiloxane resin composition for optical waveguides (2), whose cured product has a refractive index at least 0.1% higher than that of the above-mentioned composition (1), to the cured product thereof and curing it by heating, 3 applying the aforementioned composition (1) to the cured product thereof and curing it by heating, and then 4 peeling the cured product off 25 the substrate.

[0058] In this case, the cured product of the above-mentioned composition (1) is used for a bottom cladding layer and a top cladding layer, with the cured product of the abovementioned composition (2) serving as a core layer. A film-shaped channel optical waveguide is obtained by imparting a desired shape to the cured product of the abovementioned composition (2), and then coating the above-mentioned composition (1) onto the cured product of the above-mentioned composition (1) and the cured product of the above-mentioned composition (2) and curing it, followed by peeling from the substrate, if necessary.

[0059] When imparting the desired shape to the cured product of the above-mentioned composition (2), it is best to conduct etching using a desired pattern. At such time, a core pattern is formed by forming a resist pattern on the cured product of the above-mentioned composition (2) (core layer) by means of photolithography, removing the core layer that is not protected by the resist pattern by etching, and then removing said resist.

In addition, a film-shaped channel optical waveguide is also obtained by coating the above-mentioned curable organopolysiloxane resin composition for optical waveguides (1) onto a substrate and curing it by heating, peeling the cured product (bottom cladding layer) from the substrate, and forming a core pattern and a top cladding layer on top of the above-mentioned cured product in accordance with the gist of the above description.

[0060] The curable organopolysiloxane resin composition for optical waveguides used in the above-described processes is preferably liquid at normal temperature, and, in particular, has a viscosity of 20 to  $1\times10^4$  mPa·s at 25°C.

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[0061] Preferable compositions contain component (D), in particular, component (d1). Although the curing temperature used at such time depends on the components that constitute said composition, in particular, on the type and amount of component (C), and, furthermore, on the type and amount of the cure retarder, usually it is in the range of from 80 to 200°C. The process used for coating the curable organopolysiloxane resin composition for optical waveguides used in the above-mentioned fabrication processes is preferably spin coating because it allows for quickly forming uniform films. During the fabrication of film-shaped optical waveguides, solvent casting may be used instead of spin coating.

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[0062] The mold used in the second fabrication process is preferably a mold made up of a substrate whose thermal expansion coefficient is smaller than the thermal expansion coefficient of the cured product of organopolysiloxane resin. Naturally, the cured product of organopolysiloxane resin must be easy to remove from the mold. If it has insufficient mold release properties, it is better to carry out molding only after coating the inner surface of the mold with a mold release agent.

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[0063] An optical waveguide having a high transmittance in the communication wave length region can be fabricated by ① casting a curable organopolysiloxane resin composition for awaveguides into a mold having on its inner surface protrusions corresponding to the core and curing it by heating, ② the molding is removed from the mold, ③ casting a curable organopolysiloxane resin composition for optical waveguides (4), whose cured product has a refractive index at least 0.1% higher than that of the aforementioned composition (3), into the hollow portion of the cured product removed from the mold and curing it by heating, and then ④ coating the aforementioned composition (3) onto the cured product of the aforementioned composition (4) and the cured product of the aforementioned composition (3) and curing by heating.

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[0064] At such time, the cured product that is removed from the mold is fitted into another mold, after which the above-mentioned curable organopolysiloxane resin

composition for optical waveguides (4) is cast into it and cured by heating, and the above-mentioned composition (3) is coated and cured by heating on top of the cured product of the above-mentioned composition (4) and the cured product of the above-mentioned composition (3).

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[0065] The curable organopolysiloxane resin composition used in the second fabrication process preferably contains component (D) and, in particular, component (d2). Its viscosity at 25°C is preferably 20 to  $1\times10^4$  mPa·s.

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[0066] The width of the hollow portion, into which the curable organopolysiloxane resin composition is cast, is usually not more than several tens of  $\mu$ m, which is why the viscosity of the curable organopolysiloxane resin composition used for the core has to be sufficiently low to permit easy casting. Because the viscosity of said composition decreases as the temperature is raised, casting is possible if the above-mentioned viscosity at 25°C is  $1\times10^7$  mPa·s or less, but casting at the temperature range close to room temperature requires an even smaller viscosity.

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[0067] While the curing temperature at such time depends on the type and amount of the components making up said composition, in particular, component (C), and, furthermore, on the type and amount of the cure retarder, in case of a curable organopolysiloxane resin composition consisting of component (A), component (B) and component (C), the temperature is preferably in the range of from 80 to 180°C, and in case of a curable organopolysiloxane resin composition consisting of component (A), component (B), component (C), and component (D) (d2) hydrosilation reactive organosiloxane-based reactive diluent, the temperature is preferably in the range of from 20 to 180°C.

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[0068] Under heating, dimensions of the mold, which is made up of metal or ceramics with a small coefficient of thermal expansion, practically do no change, and, on the other hand, the cured product of organopolysiloxane resin, which has a large coefficient of thermal expansion, undergoes thermal expansion and is subjected to pressure as a result. Thus, the cured product of organopolysiloxane resin comes into close contact with the surface of the mold and the pattern from the surface of the mold is accurately transferred to the cured product of organopolysiloxane resin. Upon termination of curing, the mold is cooled and the cured product of organopolysiloxane resin shrinks. Slow cooling is preferable to minimize the generation of shrinkage stress due to local cooling.

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[0069] The curable organopolysiloxane resin composition for optical waveguides of the present invention has low temperature dependence of the refractive index, high heat resistance, high transparency at the telecommunication wavelength region without introducing deuterated alkyl groups and fluorinated hydrocarbon groups, possesses elasticity and hardness that makes it difficult to deform, and permits easy adjustment of

the refractive index difference between the core material and cladding material during the fabrication of an optical waveguide.

[0070] An optical waveguide comprising the cured product of the organopolysiloxane resin of the present invention have low temperature dependence of the refractive index, high heat resistance, high transparency at the telecommunication wavelength region even without the introduction of deuterated alkyl groups and fluorinated hydrocarbon groups, possess elasticity and hardness that makes them difficult to deform, and permit easy adjustment of the refractive index difference between the core material and cladding material during the fabrication of optical waveguides. In addition, an optical waveguide comprising the cured products of the organopolysiloxane resin of the present invention rarely exhibit transmission losses in the telecommunication wavelength band and are thus suitable for use as materials for optical telecommunications and optical interconnects in the near infrared region and optical integrated circuits in the near infrared region.

[0071] The process for fabricating an optical waveguide of the present invention makes it possible to easily and economically produce an optical waveguide comprising the cured product of organopolysiloxane resin that has low temperature dependence of the refractive index, high heat resistance, high transparency at the telecommunication wavelength region even without the introduction of deuterated alkyl groups and fluorinated hydrocarbon groups, possesses elasticity and hardness that makes it difficult to deform, and eliminates intermixing between the core and cladding during the fabrication of an optical waveguide.

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#### Examples

[0072] While examples and comparative examples are provided below in order to specifically explain the present invention, the present invention is not limited to these examples. Gel permeation chromatography (GPC) was used for the determination of the molecular weight of reactive diluents, organosilicon compounds, and organopolysiloxane resins used, with their number-average molecular weight calculated by comparison with a polystyrene standard. Viscosity was determined using a rotary viscometer at 25°C.

[0073] The content of silanol groups and methoxy or ethoxy groups was determined by the <sup>29</sup>Si NMR analysis. The refractive index of the cured product was determined at a wavelength of 1550 nm using a prism coupler method, and the thickness of the film was determined using Tencor Alpha-Step 200. The elastic modulus of the cured product was determined using a dynamic viscoelasticity measuring device on 2-mm thick plate, at a frequency of 1 Hz and a strain of 0.5% in the temperature range of 0 to 50°C. In addition, the transparency of the cured product was measured using a spectrophotometer with a 3-mm thick plate, at a wavelength of 1550 nm. The optical loss value at 1550 nm of the

cured product was determined by the cutback method. The polarization dependency of the cured product was observed under an optical microscope using a polarizer. The heat resistance of the cured product was evaluated by thermogravimetric analysis.

5 [0074] Summary on the organopolysiloxane resin of component (A) and organosilicon compound of component (B) used in the examples (in the formulas, Me stands for methyl, and Ph stands for phenyl);

[0075] Methylvinylphenylpolysiloxane resin (A-1):

Average unit formula: [Me<sub>2</sub>(CH<sub>2</sub>=CH)SiO<sub>1/2</sub>]<sub>0.25</sub>[PhSiO<sub>3/2</sub>]<sub>0.75</sub>, phenyl group content: 50 mol%, number-average molecular weight: 1900, viscosity: >1,000,000 mPa·s, silanol group and methoxy group content: 0.8 mol%, preparation process: 1,3-divinyl-1,1,3,3-tetramethyldisiloxane, phenyltrimethoxysilane and a catalytic amount of trifluoromethanesulfonic acid-containing water were subjected to agitation at reflux under heating to bring about hydrolytic condensation, and, after neutralizing said acid catalyst with a base, water and methanol were eluted, a small amount of potassium hydroxide was added to the residue dissolved in toluene, dehydration was carried out under heating and agitation, and, after neutralizing the potassium hydroxide with an acid, the salt was filtered off and toluene was subjected to elution.

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[0076] Methylvinylphenylpolysiloxane resin (A-2):

Average unit formula: [Me<sub>2</sub>(CH<sub>2</sub>=CH)SiO<sub>1/2</sub>]<sub>0.25</sub>[PhSiO<sub>3/2</sub>]<sub>0.65</sub>, phenyl group content: 46 mol%, number-average molecular weight: 2300, viscosity: >1,000,000 mPa·s, silanol group and ethoxy group content: 1 mol%, preparation process: vinyldimethylchlorosilane, phenyltrichlorosilane, tetraethylorthosilicate, and water were subjected to agitation at reflux under heating to bring about hydrolytic condensation, and, after neutralizing the resultant hydrochloric acid with a base, water and ethanol were eluted, a small amount of potassium hydroxide was added to the residue dissolved in toluene, dehydration was carried out under heating and agitation, and, after neutralizing the potassium hydroxide with acid, the resultant salt was filtered off and toluene was eluted.

[0077] Methylvinylphenylpolysiloxane resin (A-3):

Average unit formula: [Me<sub>2</sub>(CH<sub>2</sub>=CH)SiO<sub>1/2</sub>]<sub>0.25</sub>[MeSiO<sub>3/2</sub>]<sub>0.40</sub>[PhSiO<sub>3/2</sub>]<sub>0.35</sub>, phenyl group content: 23 mol%, number-average molecular weight: 4600, viscosity: >1,000,000 mPa·s, silanol group and methoxy group content: 1 mol%, preparation process: 1,3-divinyl-1,1,3,3-tetramethyldisiloxane, methyltrimethoxysilane, phenyltrimethoxysilane and a catalytic amount of trifluoromethanesulfonic acid-containing water were subjected to agitation at reflux under heating to bring about hydrolytic condensation, and, after neutralizing said acid catalyst with a base, water and methanol were eluted, a small amount of potassium hydroxide was added to the residue dissolved in toluene, dehydration was carried out under heating and agitation, and, after neutralizing the

and toluene, which was used as component (d1), using the proportions listed in Table 1 below (unit: g).

[0082] Using a chamber-open type system, the coating solutions were spin coated onto a silicon substrate at 2000 rpm and left stand for 10 minutes at room temperature. After that, hydrosilation-cured products of the methylphenylhydrogenpolysiloxane and the methylvinylphenylpolysiloxane resins with a uniform thickness of 7 to 8 μm were obtained by heating at 180°C for 90 minutes.

The viscosities of the coating solutions, as well as the transparency and refractive indices of the cured products of the coating solutions are listed in Table 1.

[0083] [Table 1]

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f	T		Y	T	T .
	Composition	Composition	Composition	Composition	Composition
	1	2	3	4	5
(A-1)	20.0	20.0			
(A-2)			20.0		17.0
(A-3)				20.0	3.0
(B-1)	6.4		6.9	8.1	7.1
(B-2)		6.4			
(C)	0.004	0.004	0.004	0.004	0.004
(E)	0.008	0.008	0.008	0.008	0.008
(d-1)	11.3	11.3	11.5	12.0	11.6
Viscosity-1	100,000	94,000	170,000	250,000	195,000
Viscosity-2	30	28	35	45	38
Refractive	1.530	1.526	1.525	1.500	1.519
index					
Transmissivity	96.0	95.8	95.2	94.0	94.0

(Note: "Viscosity-1" stands for the viscosity of the composition without component (d1) (unit: mPa·s)). "Viscosity-2" stands for the viscosity of the composition when it contains component (d1) (unit: mPa·s)). Both "refractive index" and "transparency" refer to values obtained by measurement of the cured products.)

[0084] All the cured products were colorless and transparent, with an elastic modulus of 0.8 to 1.0 GPa, in other words, they possessed elasticity and hardness that prevented them from being easily bent and had sufficient self-maintaining strength, and no changes due to dissolution, swelling, etc. were observed even after immersion in toluene overnight. In addition, the transmittance of the cured products in the telecommunication wavelength

region were 94% or higher, with the optical loss value being 0.5 dB/cm, i.e. sufficient for use as optical waveguides.

## Example 2

- 5 [0085] This example illustrates the formation of a channel type optical waveguide consisting of a cured product of methylvinylphenylpolysiloxane resin fabricated by coating process.
  - Using a chamber-open type system, composition 5 of Table 1 was spin coated onto a silicon substrate at 2000 rpm and left stand for 10 minutes at room temperature. After
- that, a film consisting of a hydrosilation-cured product of the methylphenylhydrogenpolysiloxane and the methylvinylphenylpolysiloxane resins with a uniform thickness of 7 μm was obtained by heating at 180°C for 90 minutes. Its refractive index was 1.519. Next, the film consisting of the cured product was used for the bottom cladding layer and composition 3 of Table 1 was spin coated on top of it at 2000 rpm and
- left stand for 10 minutes at room temperature. After that, a film consisting of a hydrosilation-cured product of the methylphenylhydrogenpolysiloxane and the methylvinylphenylpolysiloxane resins with a uniform thickness of 7  $\mu$ m was formed by heating at 180°C for 90 minutes.
- [0086] A photoresist with a thickness of 2.0 μm was coated and patterned on top of the film made of the cured product that was used as the core layer. Using the resist as a photomask, reactive ion etching was carried out using a mixed gas containing CF<sub>4</sub>, thereby creating a linear rectangular pattern of the core layer with a length of 50 mm, a width of 7.0 μm, and a height of 7.0 μm. The resist was then removed. Finally, spin coating of composition 5 of Table 1 at 2000 rpm onto the core pattern and the bottom cladding layer and subsequent heating at 180°C for 90 minutes yielded a channel type optical waveguide. In the optical waveguide, there was no intermixing between the core and cladding while the optical loss value was 0.4 dB/cm. In addition, when the optical waveguide was heated in a stream of air, no weight loss was observed up to 280°C, which confirmed its superior heat resistance.

#### Example 3

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[0087] This example illustrates the formation of a slab type optical waveguide consisting of a cured product of methylvinylphenylpolysiloxane resin fabricated by coating process.

- After coating composition 5 of Table 1 onto a glass substrate and allowing it to stand for about 30 minutes at room temperature, a film made up of a hydrosilation-cured product of the methylphenylhydrogenpolysiloxane and the methylvinylphenylpolysiloxane resins with a thickness of 50  $\mu$ m is fabricated by curing the composition by heating at 100°C for 1 hour and at 180°C for 1 hour and then precling it from the glass substrate at room
- 1 hour and at 180°C for 1 hour and then peeling it from the glass substrate at room temperature. Its refractive index was 1.519. The film made up of the cured product did

optical waveguide was heated in a stream of air, no weight loss was observed up to 280°C, which confirmed its superior heat resistance.

## Example 5

5 [0091] This example illustrates the formation of a channel type optical waveguide consisting of a cured product of methylvinylphenylpolysiloxane resin fabricated by casting process.

A colorless transparent methylvinylphenylpolysiloxane resin composition 6 was prepared by uniformly mixing 10g of methylvinylphenylpolysiloxane resin (A-2) as component (A) with 10g of methylvinylphenylsiloxane oligomer (d2) as component (d2), and then adding 13g of methylphenylhydrogenpolysiloxane (B-1) as component (B), 5 mg of a platinum/1,3-divinyltetramethyldisiloxane complex [platinum content: 2 wt%] (C) as component (C), and 10 mg of methyl(tris(1,1-dimethyl-2-propinoxy))silane (E) as a cure retarder and uniformly mixing the ingredients. The viscosity of the composition was 200 mPa·s.

[0092] A colorless transparent methylvinylphenylpolysiloxane resin composition 7 was prepared by uniformly mixing 10g of methylvinylphenylpolysiloxane resin (A-3) as component (A) with 10g of methylvinylphenylsiloxane oligomer (d2) as component (d2), and then adding 13.9g of methylphenylhydrogenpolysiloxane (B-1) as component (B), 5 mg of a platinum/1,3-divinyltetramethyldisiloxane complex [platinum content: 2 wt%] (C) as component (C), and 10 mg of methyl(tris(1,1-dimethyl-3-propinoxy))silane (E) as a cure retarder and uniformly mixing the ingredients. The viscosity of the composition was 240 mPa·s.

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[0093] With the exception of using composition 7 for the cladding and composition 6 for the core and casting and removing them from a mold at 25°C, a cord-shaped channel type optical waveguide was fabricated in the same manner as in Example 4. The refractive index of the cladding portion was 1.482 and that of the core portion was 1.505. The optical waveguide exhibited no intermixing between the core and cladding and no cracking while no cracking or peeling even after repeated bending as well as no polarization dependence were observed. When the optical waveguide was heated in a stream of air, no weight loss was observed up to 280°C, which confirmed its superior heat resistance.

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## Comparative Example 1

[0094] This example illustrates the formation of a flexible optical waveguide based on an application example of Japanese Patent Application Publication No. Sho 63-217306. To a mixture of 100 parts by weight of a linear trimethylsiloxy-terminated dimethylsiloxane-methylphenylsiloxane copolymer with a viscosity of 5000 mPa·s (25 mol% phenyl groups, refractive index: 1.50) and 5 parts by weight of a trimethylsiloxy-terminated dimethylsiloxane-methylhydrogensiloxane copolymer with a viscosity of 5

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mPa·s was added a chloroform solution of chloroplatinic acid to yield a hydrosilation-curable liquid silicone composition with a platinum content of 5 ppm by weight. The composition was extruded in a hot air form a strand-shape silicone rubber core with a diameter of 1 mm.

- 5 [0095] Subsequently, after coating a hydrosilation-curable liquid silicone rubber composition with a viscosity of 3000 mPa·s (main ingredients: linear dimethylvinylsiloxy-terminated dimethylpolysiloxane, trimethylsiloxy-terminated dimethylsiloxane-methylhydrogensiloxane copolymer, and chloroplatinic acid, refractive index: 1.41) on the core, a strand-shaped flexible optical waveguide with a diameter of 1.4 mm was fabricated by curing the composition by heating it for 5 minutes at 150°C. The optical waveguide could be easily deformed and had no self-maintaining strength. In addition, when it was heated in the air, a weight loss due to decomposition was observed starting from 200°C, which indicated inferior heat resistance.
- Comparative Example 2
  [0096] This example illustrates the formation of a polysilsesquioxane optical waveguide based on Working Example 2 of Japanese Patent Application Publication No. Hei 4-157402.
- Using polymethylsilsesquioxane (molecular weight Mw=2000, refractive index: 1.423)
  for the cladding and polyphenylsilsesquioxane (molecular weight Mw=15000, refractive index: 1.555) for the core, a channel type optical waveguide, in which the core made of polyphenylsilsesquioxane and had a with of 8 μm, a height of 8 μm, and a length of 50 mm and both the top cladding layer made of polymethylsilsesquioxane and bottom cladding layer made of polymethylsilsesquioxane had a thickness of 20 μm, was
  fabricated by spin coating, photolithography, dry etching, and then again using spin coating as the final step. The optical loss value of the optical waveguide was not more than 0.5 dB/cm. When the optical waveguide was subjected to heat treatment at 260°C for 2 minutes, the refractive index of the core decreased by 0.3% and the refractive indices of both cladding layers decreased by 0.8%, thus changing the refractive index difference
  between the two in comparison with the difference that existed prior to the heat treatment
- between the two in comparison with the difference that existed prior to the heat treatment. This indicated that this optical waveguide had inferior heat resistance.

## **Industrial Applicability**

[0097] The curable organopolysiloxane resin composition for optical waveguides of the present invention is useful for fabricating various optical waveguides and is suitable for use as materials for optical communications and optical integrated circuits in the near infrared region, the optical waveguide of the present invention is useful as passive optical waveguide exemplified by non-branching waveguides, branching waveguides, optical splitters, and optical couplers, and active optical waveguide exemplified by waveguide-type optical switches, waveguide-type optical modulator, optical attenuators, and optical amplifiers, and the process for fabricating an optical waveguide of the present invention is useful for fabricating various optical waveguides with lower process cost.

- 1. (amended) A curable organopolysiloxane resin composition for optical waveguides comprising (A) an organopolysiloxane resin, which is represented by the average unit formula (1):
- (R¹₃SiO₁₂)a(R²₂SiO₂₂)b(R³SiO₃₂)c(SiO₄₂)d
  (wherein R¹, R², and R³ stand for one, two, or more kinds of monovalent hydrocarbon groups selected from monovalent aliphatic hydrocarbon groups having 1 to 6 carbon atoms and monovalent aromatic hydrocarbon groups having 6 to 10 carbon atoms, 0<a≤0.5, 0≤b<0.2, 0.3≤c<1, 0≤d≤0.4, 0≤(b+d)/(a+c)≤0.25, and a+b+c+d=1)and has three</li>
  or more monovalent unsaturated aliphatic hydrocarbon groups per molecule, with not less than 10 mol% of the monovalent hydrocarbon groups being monovalent aromatic hydrocarbon groups, (B) an organosilicon compound having two or more silicon-bonded hydrogen atoms per molecule, with not less than 5 mol% of all the silicon-bonded monovalent substituent groups being monovalent aromatic hydrocarbon groups, and (C) a hydrosilation catalyst.
  - 2. (amended) The curable organopolysiloxane resin composition for optical waveguides according to claim 1, wherein the viscosity of the composition is not more than  $1\times10^7$  mPa·s at 25°C.
- (R<sup>1</sup><sub>3</sub>SiO<sub>1/2</sub>)<sub>a</sub>(R<sup>2</sup><sub>2</sub>SiO<sub>2/2</sub>)<sub>b</sub>(R<sup>3</sup>SiO<sub>3/2</sub>)<sub>c</sub>(SiO<sub>4/2</sub>)<sub>d</sub> (1)
  (wherein R<sup>1</sup>, R<sup>2</sup>, and R<sup>3</sup>, a, b, c, d, (b+d)/(a+c), and a+b+c+d are the same as above) and has three or more monovalent unsaturated aliphatic hydrocarbon groups per molecule, with not less than 10 mol% of the monovalent hydrocarbon groups being monovalent aromatic hydrocarbon groups, (B) an organosilicon compound having two or more silicon-bonded hydrogen atoms per molecule, with not less than 5 mol% of all the
  silicon-bonded monovalent substituent groups being monovalent aromatic hydrocarbon groups, (C) a hydrosilation catalyst, and (D) (d1) a solvent or (d2) a hydrosilation-
- 4. (amended) An optical waveguide comprising a hydrosilation-cured product of (A) an organopolysiloxane resin, which is represented by the average unit formula (1): (R¹₃SiO₁₂₂)a(R²₂SiO₂₂₂)b(R³SiO₃₂₂)c(SiO₄₂₂)d (1) (wherein R¹, R², and R³ stand for one, two, or more kinds of monovalent hydrocarbon groups selected from monovalent aliphatic hydrocarbon groups having 1 to 6 carbon atoms and monovalent aromatic hydrocarbon groups having 6 to 10 carbon atoms, 0<a≤0.5, 0≤b<0.2, 0.3≤c<1, 0≤d≤0.4, 0≤(b+d)/(a+c)≤0.25, and a+b+c+d=1) and has</li>

three or more monovalent unsaturated aliphatic hydrocarbon groups per molecule, with

reactive organosiloxane-based diluent.

not less than 10 mol% of the monovalent hydrocarbon groups being monovalent aromatic hydrocarbon groups, and (B) an organosilicon compound having two or more siliconbonded hydrogen atoms per molecule, with not less than 5 mol% of all the silicon-bonded monovalent substituent groups being monovalent aromatic hydrocarbon groups.

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- 5. (amended) An optical waveguide comprising a hydrosilation-cured product of (A) an organopolysiloxane resin, which is represented by the average unit formula (1):  $(R^1{}_3SiO_{1/2})_a(R^2{}_2SiO_{2/2})_b(R^3SiO_{3/2})_c(SiO_{4/2})_d \qquad \qquad (1)$  (wherein  $R^1$ ,  $R^2$ ,  $R^3$ , a, b, c, d, (b+d)/(a+c), and a+b+c+d are the as described above) and has three or more monovalent unsaturated aliphatic hydrocarbon groups per molecule, with not less than 10 mol% of the monovalent hydrocarbon groups being monovalent aromatic hydrocarbon groups, (B) an organosilicon compound having two or more
- silicon-bonded hydrogen atoms per molecule, with not less than 5 mol% of all the silicon-bonded monovalent substituent groups being monovalent aromatic hydrocarbon groups, and (d2) a hydrosilation-reactive organosiloxane-based diluent.
  - 6. (cancelled)
- 20 7. (cancelled)
  - 8. (amended) The optical waveguide according to claim 4 wherein both the cladding and the core of the optical waveguide consist of a hydrosilation-cured product of component (A) and component (B), with the refractive index of the core being at least 0.1% higher than the refractive index of the cladding.
    - 9. (amended) The optical waveguide according to claim 5, wherein both the cladding and the core of the optical waveguide consist of a hydrosilation-cured product of component (A), component (B), and component (d2), with the refractive index of the core being at least 0.1% higher than the refractive index of the cladding.
    - 10. (amended) The optical waveguide according to claim 8, wherein the refractive index difference is regulated by making the total content of monovalent aromatic hydrocarbon groups in component (A) and component (B) used for the core higher than the total content of monovalent aromatic hydrocarbon groups in component (A) and component (B) used for the cladding.
- 11. (amended) The optical waveguide according to claim 9, wherein the refractive 40 index difference is regulated by making the total content of monovalent aromatic hydrocarbon groups in component (A), component (B), and component (d2) used for the

core higher than the total content of monovalent aromatic hydrocarbon groups in component (A), component (B), and component (d2) used for the cladding.

- 12. (amended) The optical waveguide according to any of claims 4, 5, 8 to 11, whichhas a film-like shape.
  - 13. (amended) A process for fabricating an optical waveguide, wherein the curable organopolysiloxane resin composition for optical waveguides s according to any of claim 1 to claim 3 is cured by heating.
  - 14. (amended) A process for fabricating an optical waveguide, wherein the curable organopolysiloxane resin composition for optical waveguides according to any of claim 1 to claim 3 is applied to a substrate and cured by heating.
- 15. (amended) A process for fabricating a slab optical waveguide, in which a curable organopolysiloxane resin composition for optical waveguides (1) according to any of claim 1 to claim 3 is applied to a substrate and cured by heating, a curable organopolysiloxane resin composition for optical waveguides (2), whose cured product has a refractive index at least 0.1% higher than that of the above-mentioned composition
  (1), is applied to the cured product thereof and cured by heating, whereupon the aforementioned composition (1) is applied to the cured product thereof and cured by heating.
- 16. (amended) A process for fabricating an optical waveguide, wherein the curable
   organopolysiloxane resin composition for optical waveguides according to any of claim 1 to claim 3 is casted into a mold having a desired inner surface shape and cured by heating.
- organopolysiloxane resin composition for optical waveguides (3) according to any of claim 1 to claim 3 is cast into a mold having on its inner surface protrusions corresponding to the core and cured by heating, ② the molding is removed from the mold, ③ a curable organopolysiloxane resin composition for optical waveguides (4) according to any of claim 1 to claim 3, whose cured product has a refractive index at least 0.1% higher than that of the aforementioned composition (3), is cast into the hollow portion of the cured product removed from the mold and cured by heating, whereupon ④ the aforementioned composition (3) is applied on top of the cured product of the aforementioned composition (4) and the cured product of the aforementioned composition (3) and cured by heating.